FORM PTO-1390 (REV. 11-2000) U S DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE

TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371

ATTORNEY 'S DOCKET NUMBER: 1-15478

U S APPLICATION NO (If known, see 37 CFR 1 5): 09/914082

INTERNATIONAL APPLICATION NO.	INTERNATIONAL FILING DATE	PRIORITY DATE CLAIMED
PCT/DE00/00553	24 February 2000 (24.02.00)	01 March 1999 (01.03.99)

TITLE OF INVENTION: ELECTROCHROMIC ELEMENT

APPLICANT(S) FOR DO/EO/US: DIRK JOEDICKE et al.

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. [XX] This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. [ ] This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. [X] This is an express request to begin national examination procedures (35 U.S.C. 371(f)). The submission must include items (5), (6), (9) and (21) indicated below.
4. [X] The US has been elected by the expiration of 19 months from the priority date (Article 31).
5. [X] A copy of the International Application as filed (35 U.S.C. 371(c)(2))
   a. [ ] is attached hereto (required only if not communicated by the International Bureau).
   b. [X] has been communicated by the International Bureau.
   c. [ ] is not required, as the application was filed in the United States Receiving Office (RO/US).
6. [X] An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)).
   a. [XX] is attached hereto.
   b. [ ] has been previously submitted under 35 U.S.C. 154(d)(4).
7. [ ] Amendments to the claims of the International Aplication under PCT Article 19 (35 U.S.C. 371(c)(3))
   a. [ ] are attached hereto (required only if not communicated by the International Bureau).
   b. [ ] have been communicated by the International Bureau.
   c. [ ] have not been made; however, the time limit for making such amendments has NOT expired.
   d. [ ] have not been made and will not be made.
8. [ ] An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371 (c)(3)).
9. [X] An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).
10. [ ] An English lanugage translation of the annexes of the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).

Items 11 to 20 below concern document(s) or information included:

11. [ ] An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
12. [ ] An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
13. [X] A FIRST preliminary amendment.
14. [ ] A SECOND or SUBSEQUENT preliminary amendment.
15. [ ] A substitute specification.
16. [ ] A change of power of attorney and/or address letter.
17. [ ] A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821 - 1.825.
18. [ ] A second copy of the published international application under 35 U.S.C. 154(d)(4).
19. [ ] A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4).
20. [X] Other items or information: Express Mail Certificate, return card, 1 sheet formal drawings; copy of cover page of published Int'l Appln; Copy of search report ISA/210 with English translation; Copy of IPEA Report

U.S. APPLICATION NO (if known, see 37 CFR 1.5)	INTERNATIONAL APPLICATION NO	ATTORNEY'S DOCKET NUMBER
09/914082	PCT/DE00/00553	1-15478

21. [X] The following fees are submitted:

BASIC NATIONAL FEE (37 CFR 1.492 (a) (1) - (5)):

		CALCULATIONS PTO USE ONLY
Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO	**$1000.00**	
International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO	**$860.00**	
International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO	**$710.00**	
International preliminary examination fee (37 CFR 1.482) paid to USPTO but all claims did not satisfy provisions of PCT Article 33(1)-(4)	**$690.00**	
International preliminary examination fee (37 CFR 1.482) paid to USPTO and all claims satisfied provisions of PCT Article 33(1)-(4)	**$100.00**	
ENTER APPROPRIATE BASIC FEE AMOUNT =		$ 860.00
Surcharge of **$130.00** for furnishing the oath or declaration later than [ ] 20 [ ] 30 months from the earliest claimed priority date (37 CFR 1.492(e)).		$ -

CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE	
Total claims	14 - 20 =	0	x **$18.00**	$ -
Independent claims	1 - 3 =	0	x **$80.00**	$ -
MULTIPLE DEPENDENT CLAIM(S) (if applicable)			+ **$270.00**	$ -
TOTAL OF ABOVE CALCULATIONS =				$ 860.00
[ ] Applicant claims small entity status. See 37 CFR 1.27. The fees indicated above are reduced by 1/2.			+	$ -
SUBTOTAL =				$ 860.00
Processing fee of **$130.00** for furnishing the English translation later than [ ] 20 [ ] 30 months from the earliest claimed priority date (37 CFR 1.492(f)).				$ -
TOTAL NATIONAL FEE =				$ 860.00
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). **$40.00** per property +				$ -
TOTAL FEES ENCLOSED =				$ 860.00
			Amount to be refunded:	$
			charged:	$

a. [X] A check in the amount of $ 860.00 to cover the above fees is enclosed.

b. [ ] Please charge my Deposit Account No. _____ in the amount of $ _____ to cover the above fees. A duplicate copy of this sheet is enclosed.

c. [X] The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 13-1816. ~~A duplicate copy of this sheet is enclosed.~~

d. [ ] Fees are to be charged to a credit card. **WARNING:** Information on this form may become public. ***Credit card information should not be included on this form.*** Provide credit card information and authorization on PTO-2038.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137 (a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO
Marshall & Melhorn, LLC
Phillip S. Oberlin
Four SeaGate - 8th Floor
Toledo, Ohio 43604
Phone: (419) 249-7149
Fax: (419) 249-7151

SIGNATURE

PHILLIP S. OBERLIN
NAME

19,066
REGISTRATION NUMBER

09914082 .030102

"Express Mail" Label Number EL 469907073 US

I hereby certify that this correspondence is being deposited with the United States Postal Service "Express Mail Post Office to Addressee" service under 37 CFR 1.10 on March 11, 2002 and is addressed to the Commissioner for Patents, Box PCT, U.S. Patent and Trademark Office, Washington, D C 20231.

Kathleen J. Moore
(Signature of person mailing correspondence)

Kathleen J Moore
(Typed name of person mailing correspondence)

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of: DIRK JOEDICKE, HANS-JOACHIM BECKER, VOLKER GRUMPICH and RICHARD BATCHELOR	]	Group Art Unit:
Serial No. 09/914,082 filed	]	Examiner:
Filing Under 35 U.S.C. 371 in the DO/EO/US off PCT/DE00/00553 filed 24 February 2000	]	
For: ELECTROCHROMIC ELEMENT	]	Attorney Docket: 1-15478

March 11, 2002

Commissioner for Patents
Box PCT
United States Patent and Trademark Office
Washington, D.C. 20231

APPENDIX E
SECOND PRELIMINARY AMENDMENT

Honorable Sir:

In respect of the above-entitled application, please amend the designation of joint inventors, as follows:

4-00 Add RICHARD BATCHELOR, as co-inventor.

Respectfully submitted,

Donald A. Schurr
Registration No. 34,247

ATTORNEYS
MARSHALL & MELHORN, LLC
Four SeaGate - 8th Floor
Toledo, Ohio 43604

"Express Mail" Label Number EL 469907073 US
I hereby certify that this correspondence is being deposited with the United States Postal Service "Express Mail Post Office to Addressee" service under 37 CFR 1.10 on March 11, 2002 and is addressed to the Commissioner for Patents, Box PCT, U S. Patent and Trademark Office, Washington, D.C. 20231.

Kathleen J. Moore
(Signature of person mailing correspondence)
Kathleen J. Moore
(Typed name of person mailing correspondence)

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of: DIRK JOEDICKE, HANS-JOACHIM BECKER, VOLKER GRUMPICH and RICHARD BATCHELOR	Group Art Unit:
Serial No. 09/914,082	Examiner:
Filing Under 35 U.S.C. 371 in the DO/EO/US off PCT/DE00/00553 filed 24 February 2000	
For: ELECTROCHROMIC ELEMENT	Attorney Docket: 1-15478

March 11, 2002

Commissioner for Patents
Box PCT
United States Patent and Trademark Office
Washington, D.C. 20231

REQUEST FOR CORRECTION OF INVENTOR'S ADDRESS

Honorable Sir:

We have been informed that the address of the third inventor in the above-captioned application has changed as follows:

	Old address	New address
VOLKER GUMPRICH: 3-00	Thuermchenswall 56/58 D-50668 Koeln Germany	Berghauschensweg 242 D-41468 Neuss Germany

Applicants respectfully request that this change of address be noted and made of record in the captioned application.

Respectfully submitted,

Donald A. Schurr
Registration No. 34,247

ATTORNEYS
Marshall & Melhorn, LLC
Four SeaGate - 8th Floor
Toledo, Ohio 43604

1-15478

"Express Mail" Label Number EL850046470US

I hereby certify that this correspondence is being deposited with the United States Postal Service "Express Mail Post Office to Addressee" service under 37 CFR 1.10 on August 22, 2001 and is addressed to the Commissioner for Patents, Box PCT, Washington, D.C. 20231.

Kathleen J. Moore
(signature of person mailing correspondence)

Kathleen J. Moore
(Typed name of person mailing correspondence)

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of: DIRK JOEDICKE, HANS-JOACHIM BECKER and VOLKER GUMPRICH	Group Art Unit:
Serial No. Filed:	Examiner:
Filing Under 35 U.S.C. 371 in the DO/EO/US off PCT/DE00/00553 filed 24 February 2000	
For: ELECTROCHROMIC ELEMENT	Attorney Docket 1-15478

August 22, 2001

Box PCT
Commissioner for Patents
Washington, D.C. 20231

PRELIMINARY AMENDMENT

Honorable Sir:

Prior to the first Office Action and before calculating the filing fee, please amend the application being filed concurrently herewith under 35 U.S.C. 371 as follows:

In the Specification:

Page 1, above line 1, insert --TITLE--; line 2, insert --BACKGROUND OF THE INVENTION--.

Page 3, line 6, insert --SUMMARY OF THE INVENTION--.

Page 7, line 28, insert --BRIEF DESCRIPTION OF THE DRAWINGS--.

Page 8, line 6, insert --DESCRIPTION OF THE PREFERRED EMBODIMENT--.

In the Claims:

Please amend original claims 1-12 as follows:

1. (Amended) An electrochromic element with an electrochromic arrangement enclosed between two plane substrates, which comprises at least two electrode layers, one electrochromic layer, one ion storage layer, and one polymer electrolyte layer formed in situ, where the polymer electrolyte layer adjoins a sealing element at the edge of the electrochromic element, wherein the sealing element comprises a plastically deformable liquid impermeable adhesive strip of a polyacrylate, arranged between the two plane substrates and adjoining directly the polymer electrolyte layer, as well as of a sealing strand adjacent thereto on the outside, comprising a gas impermeable sealant chemically compatible with the adhesive strip.

2. (Amended) An electrochromic element according to Claim 1, wherein the adhesive strip is formed of a polyacrylate tape.

3. (Amended) An electrochromic element according to Claim 1, wherein the adhesive strip possesses a width of at least 5 mm.

4. (Amended) An electrochromic element according to Claim 3, wherein the adhesive strip possesses a maximum width of 20 mm.

5. (Amended) An electrochromic element according to Claim 1, wherein the adhesive strip comprises a polyacrylate with a maximum water content of 0.3 weight percent, preferably less than 0.05 weight percent.

6. (Amended) An electrochromic element according to Claim 1, wherein the adhesive strip comprises a polyacrylate with a glass transition temperature below 10°C.

7. (Amended) An electrochromic element according to claim 1, wherein the sealing strand comprises a polyisobutylene or butyl rubber based butyl sealant.

8. (Amended) An electrochromic element according to Claim 7, wherein the sealing strand possesses a specific conductivity of less than $10^{-9}\ \Omega^{-1}\cdot cm^{-1}$, and a water vapor permeability according to DIN 53122-1.2 of less than 0.5 $g\cdot m^{-2}\cdot d^{-1}$.

9. (Amended) An electrochromic element according to Claim 1, wherein the sealing strand comprises an epoxy sealant.

10. (Amended) An electrochromic element according to Claim 9, wherein the sealing strand possesses a specific conductivity of less than 10^{-11} $\Omega^{-1} \cdot cm^{-1}$, and a water vapor permeability according to DIN 53122-1.2 of less than 4.0 $g \cdot m^{-2} \cdot d^{-1}$.

11. (Amended) An electrochromic element according to Claim 1, wherein the sealing strand is adjoined by at least one further sealant strand, in particular one with polysulfide base.

12. (Amended) An electrochromic element according to Claim 1, wherein the polymer electrolyte layer comprises at least one (meth)acrylic ester, at least one plasticizer and at least one polymerization initiator.

Please add the following new claims:

13. An electrochromic element according to Claim 8, wherein the sealing strand possesses a specific conductivity of less than 10^{-11} $\Omega^{-1} \cdot cm^{-1}$.

14. An electrochromic element according to Claim 10, wherein the sealing strand possesses a specific conductivity of less than 10^{-13} $\Omega^{-1} \cdot cm^{-1}$.

REMARKS

Applicant has amended the specification and claims to eliminate multiple dependencies and adapt the specification and claims to U.S. patent practice. A separate marked-up version of the change made to the claims is attached hereto entitled "Version With Markings To Show Changes Made".

Claims 1-12 have been amended. New claims 13 and 14 have been added. Claims 1-14 are currently pending in the present application. No new matter has been added by any of these amendments.

Favorable consideration of the application as amended is respectfully requested.

Respectfully submitted,

Phillip S. Oberlin
Registration No. 19,066

ATTORNEYS
MARSHALL & MELHORN, LLC
Four SeaGate - 8th Floor
Toledo, Ohio 43604
Phone: (419) 249-7145
Fax: (419) 249-7151

Version With Markings To Show Changes Made

1. (Amended) [Electrochromic] An electrochromic element with an electrochromic arrangement enclosed between two plane substrates, which comprises at least two electrode layers, one electrochromic layer, one ion storage layer, and one polymer electrolyte layer formed in situ, where the polymer electrolyte layer adjoins a sealing element at the edge of the electrochromic element[. characterized in that] , wherein the sealing element [consists of] comprises a plastically deformable liquid impermeable adhesive strip [(8)] of a polyacrylate, arranged between the two plane substrates [(1, 2)] and adjoining directly the polymer electrolyte layer [(7)], as well as of a sealing strand [(9)] adjacent thereto on the outside, [consisting of] comprising a gas impermeable sealant chemically compatible with the adhesive strip [(8)].

2. (Amended) [Electrochromic] An electrochromic element [in accordance with] according to Claim 1, [characterized in that] wherein the adhesive strip [(8)] is formed of a polyacrylate tape.

3. (Amended) [Electrochromic] An electrochromic element [in accordance with] according to Claim 1 [or 2, characterized in that], wherein the adhesive strip [(8)] possesses a width of at least 5 mm.

4. (Amended) [Electrochromic] An electrochromic element [in accordance with] according to Claim 3, [characterized in that] wherein the adhesive strip [(8)] possesses a maximum width of 20 mm.

5. (Amended) [Electrochromic] An electrochromic element [in accordance with one of the foregoing claims, characterized in that] according to Claim 1, wherein the adhesive strip [(8) consists] comprises of a polyacrylate with a maximum water content of 0.3 weight percent, preferably less than 0.05 weight percent.

6. (Amended) [Electrochromic] An electrochromic element [in accordance with one of the foregoing claims, characterized in that] according to Claim 1, wherein the adhesive strip [(8) consists of] comprises a polyacrylate with a glass transition temperature below 10°C.

7. (Amended) [Electrochromic] An electrochromic element [in accordance with one of the foregoing claims, characterized in that] according to Claim 1, wherein the sealing strand [(9) consists of] comprises a polyisobutylene or butyl rubber based butyl sealant.

8. (Amended) [Electrochromic] An electrochromic element [in accordance with] according to Claim 7, [characterized by the fact that] wherein the sealing strand [(9)] possesses a specific conductivity of less than $10^{-9}\ \Omega^{-1}\cdot cm^{-1}$, [preferably less than $10^{-11}\ \Omega^{-1}\cdot cm^{-1}$] and a water vapor permeability according to DIN 53122-1.2 of less than 0.5 $g\cdot m^{-2}\cdot d^{-1}$.

9. (Amended) [Electrochromic] An electrochromic element [in accordance with one of Claims 1 to 6, characterized in that] according to Claim 1, wherein the sealing strand [(9) consists of] comprises an epoxy sealant.

10. (Amended) [Electrochromic] An electrochromic element [in accordance with] according to Claim 9, [characterized in that] wherein the sealing strand [(9)] possesses a specific conductivity of less than 10^{-11} $\Omega^{-1} \cdot cm^{-1}$, [preferably less than 10^{-13} $\Omega^{-1} \cdot cm^{-1}$,] and a water vapor permeability according to DIN 53122-1.2 of less than 4.0 $g \cdot m^{-2} \cdot d^{-1}$.

11. (Amended) [Electrochromic] An electrochromic element [in accordance with one of the foregoing claims, characterized in that] according to Claim 1, wherein the sealing strand [(9)] is adjoined by at least one further sealant strand [(10)], in particular one with polysulfide base.

12. (Amended) [Electrochromic] An electrochromic element [in accordance with one of the foregoing claims, characterized in that] according to Claim 1, wherein the polymer electrolyte layer [(7)] comprises at least one (meth)acrylic ester, at least one plasticizer and at least one polymerization initiator.

1/prt

Electrochromic Element

The invention concerns an electrochromic element with an electrochromic arrangement enclosed between two plane substrates, which arrangement comprises at least two electrode layers, one electrochromic layer, one ion storage layer, and one polymer electrolyte layer formed in situ, where the polymer electrolyte layer at the edge of the electrochromic element adjoins a sealing element.

Electrochromic elements of the aforementioned construction are known from numerous publications. They are employed inter alia for displays, dimmable mirrors, and glazing systems with variable light transmission. The plane substrates, which can be flat or also curved, consist in any case, in the case of large-area glazing systems, mostly of inorganic glass panes. They can also however consist of other materials, for example plastics. For the long-term stability of the electrochromic element it is indispensable for the substrates to be sufficiently impermeable to substances present in the environment, in particular to gases present in the ambient air. They must in addition reliably protect the electrochromic arrangement from migration of volatile constituents, such as for example plasticizers, solvents, etc. These requirements are particularly satisfactorily fulfilled by inorganic glass panes. For the sake of simplicity therefore, in the following text in connection with plane substrates, glass panes will principally be involved, without of course the invention being restricted thereto.

At least one of the electrode layers must be light-transmitting. Usually, one employs for both electrode layers transparent conductive metal oxide layers (TCOs), for example of ITO or doped tin oxide. The electrode layers have the purpose of permitting application to the electrochromic arrangement of an electric voltage, with which its light transmission can be varied. For the electrochromic layer it is usual to employ tungsten oxide-based materials, whose light transmission can be varied by embedding cations, such as H^+, Li^+, Na^+. As counterpart, the electrochromic layer requires an ion storage layer, also termed counter-electrode, for which numerous materials are known, including in particular cerium-titanium oxide and vanadium-titanium oxide.

The electrochromic layer and the ion storage layer are separated by an ion conduction layer ensuring ion transport between the two. Particularly for large-area electrochromic elements, the use of polymer electrolyte layers formed in situ has proved useful. The polymer electrolyte layers are formed by a liquid mixture containing inter alia monomers and at least one conduction salt being applied (injected) between the glass panes and being polymerized there. The polymer electrolyte layers can be present in the complete electrochromic element in solid, gel or also liquid form. They adjoin a sealing element at the edge of the electrochromic element.

The edge-side sealing element has primarily the function of sealing the electrochromic arrangement arranged between the glass panes also at the edge of the electrochromic element permanently against liquids and gases. In particular, it is necessary to prevent oxygen or water vapour present in the ambient air penetrating into the system. Within the scope of manufacture of the electrochromic element, the sealing element also performs the function of occluding the liquid monomer blend introduced between the glass panes in a liquid-impermeable manner. Such monomer blends frequently possess very low viscosity which may be below that of water, so that even minor leaks or pores in the sealing element would lead to undesirable escape of the monomer blend. Finally, the sealing element serves as a spacer between the glass panes, before the polymer electrolyte layer has hardened enough to assume this function itself.

From EP 0 836 932 A1, an electrochromic element with the features of the preamble is known, where a sealing element directly adjoins the polymer electrolyte layer, which sealing element consists of a liquid and gas-impermeable sealant which does not chemically react with the components of the electrochromic arrangement, in particular with the polymer electrolyte layer. As being suitable for this purpose, polyisobutylene-based butyl sealants are mentioned, these possessing an especially high level of impermeability to gas diffusion. The sealing element is not arranged between the glass panes, but adjoins a step formed by the glass panes and the electrochromic arrangement at the edge. With such an arrangement of the sealing element, production of the polymer electrolyte layer is rendered at least difficult. In addition, trials of the applicant have shown that sealing elements of butyl sealants during the course of polymerization of the polymer

electrolyte layer become detached from it at least locally, which can lead to premature ageing in these areas. In addition, it is extremely difficult to find a suitable sealant which is so readily compatible with the materials used for the polymer electrolyte layer that, even after a prolonged period of time, no material substitution or even a(n) (electro-) chemical reaction takes place between the two materials.

It is the object of the invention to configure electrochromic elements of the aforementioned construction so as to be permanently gas-impermeable, where undesirable interactions of the sealing element with the polymer electrolyte layer formed in situ or with other components of the electrochromic arrangement are to be prevented. Peeling of the sealing element off the polymer electrolyte layer during its polymerization should be prevented. The sealing element should, in addition, permit production of the electrochromic element, in particular introduction of the monomer blend necessary for formation of the polymer electrolyte layer, by customary production processes and reliably keep the plane substrates at a distance, at least during the production process.

This object is solved by an electrochromic element with the features of Claim 1. Advantageous configurations are the subject of the subclaims.

According to the invention, provision is made for the sealing element to consist of a plastically deformable liquid-impermeable adhesive strip of a polyacrylate directly contiguous to the polymer electrolyte layer and to the surfaces facing one another of the two plane substrates (glass panes), as well as of a sealing strand outwardly adjacent thereto consisting of a gas-impermeable sealant which is chemically compatible with the adhesive strip.

Surprisingly, it has proved successful, through the use of a sealing element according to the invention consisting of at least two separate functional components, to provide all the functions necessary for production and durable operation of an electrochromic element. Here, the adhesive strip primarily assumes the function of an edge closure for the polymer electrolyte layer, preventing escape of the liquid monomer blend, and in addition acts as a spacer for the glass panes.

By virtue of its plasticity, the adhesive strip can follow the polymer electrolyte layer contracting during the course of polymerization and does not peels off from it, as is the case with other known sealing elements. Polyacrylate materials are readily compatible with a series of materials used for the polymer electrolyte layers, in particular however with (meth)acrylic ester-based polymer electrolyte layers, which have proved especially suitable for this purpose.

The polyacrylate adhesive strip according to the invention is impermeable to liquid, but not however gas-impermeable. In particular, it is not sufficiently diffusion-impermeable to gases, such as oxygen and water vapour, and to solvents, such as propylene carbonate or ethylene carbonate. The adhesive strip is therefore, within the scope of the invention, supplemented functionally by a sealing strand of a highly gas-impermeable sealant which is chemically readily compatible with the adhesive strip.

It has been found that polyacrylate adhesive strips are especially suitable as a kind of resilient pad or adapter between the materials of the polymer electrolyte layer on the one hand and the sealants of the sealing strand on the other hand. The presence of such a resilient pad which is compatible with both materials permits the use of a wider variety of sealants than if they would adjoin directly the polymer electrolyte layer and would have to be (electro-) chemically compatible with it.

Preferably, the adhesive strip consists of adhesive tape, available in roll form, of a polyacrylate which possesses a glass transition temperature of less than 20 °C. Preferably, the glass transition temperature should be significantly below 20 °C, in particular below 10 °C.

Such adhesive tapes are for example manufactured and marketed under the commercial names Scotch Acrylic Foam or Scotch Isotact by Messrs 3M. Such adhesive tapes can be readily applied at room temperature. They adapt well to the shape of the adjacent surfaces and possess a type of self-healing effect, so that punctures which occur on penetration of the injection tools for insertion of the monomer blend between the glass panes quickly reclose of their own accord after removal of the tools. It is additionally recommended, when selecting the material of the adhesive

tape, that one should ensure that the material possesses a low to medium degree of crosslinking. This will ensure that the tackiness of the material is sufficiently high. The adhesive tape is applied prior to assembly of the components of the electrochromic element either manually or mechanically to the edges of the glass panes. Here, it is preferable to use a transparent adhesive tape. As the edge area of the electrochromic element is otherwise masked by the frame, it is also possible to use colored or opaque adhesive tapes.

Adhesive strips of polyacrylate whose water content is a maximum of 0,3 weight percent are especially suitable for durable functioning of the electrochromic elements. Preferably, the water content should be below 0.05 weight percent. If necessary, the adhesive strip should be subjected to suitable drying treatment prior to use. With a higher water content of the adhesive strip, there is the risk of diffusion of this water into the polymer electrolyte layer, which can lead to blistering and premature ageing.

The adhesive strip should preferably possess a width of at least 5 mm and a maximum of 20 mm. In the case of lesser widths, handling and achieving impermeability to liquid will be made difficult, whilst in the case of greater widths, the ratio of usable area to the overall area of the electrochromic element will be less favorable, without its properties being improved any further.

For the sealing strand, it is preferable to use a highly gas-impermeable butyl sealant of polyisobutylene or butyl rubber base, or an epoxy sealant.

Materials are preferred which, for prevention of leakage currents in the case of butyl sealants, possess a specific electrical conductivity of less than $10^{-9}\ \Omega^{-1}cm^{-1}$, preferably less than $10^{-11}\ \Omega^{-1}cm^{-1}$, and in the case of epoxy sealants a specific electrical conductivity of less than $10^{-12}\ \Omega^{-1}cm^{-1}$, preferably less than $10^{-13}\ \Omega^{-1}cm^{-1}$. In both cases, the water-vapour permeability in accordance with DIN 53122-1.2 (corresponding to prEN 1279-4) should be maximum approximately 4.0 $g \cdot m^{-2} \cdot d^{-1}$. The water-vapour permeability is determined in accordance with the Standard on 2 mm thick films from the corresponding material.

The aforementioned sealant materials are chemically especially compatible with the polyacrylate materials used for the adhesive strip according to the invention and possess excellent impermeability to diffusion in respect of gases such as oxygen and water vapour, as well as solvents such as for example polypropylene carbonate or ethylene carbonate. Other materials being common and well-known as sealants can also be used provided that they guarantee imperviousness to the gases stated comparable to the materials preferred within the scope of the invention and are compatible with the polyacrylate adhesive strips. This can be established by the specialist by means of simple tests.

Preferred butyl sealants are for example Bostik 5124 or 5125 (Messrs Bostik) with a butyl rubber base, possessing the following properties:

water-vapour permeability	approximately 0.10 - 0.15 $g \cdot m^{-2} \cdot d^{-1}$
specific electrical conductivity	approximately 10^{-11} $\Omega^{-1}cm^{-1}$

Preferred epoxy sealants are for example Araldit 2012 or 2014 (Ciba-Geigy) with the following properties:

water-vapour permeability	approximately 4.0 $g \cdot m^{-2} \cdot d^{-1}$
specific electrical conductivity	approximately 10^{-14} $\Omega^{-1}cm^{-1}$

or Eccobond 45 (Grace Specialty Polymers / Emerson & Cuming) with the following properties:

water-vapour permeability	approximately 2.5 $g \cdot m^{-2} \cdot d^{-1}$
specific electrical conductivity	approximately $3 \cdot 10^{-14}$ $\Omega^{-1}cm^{-1}$

Of course, a sealant, in particular with a base of butyl rubber, polyisobutylene or epoxy resin, functioning as edge seal of an electrochromic unit must be electrochemically inert in the electrical voltage range necessary for the function of the electrochromic element. This means that the

sealant may not evidence any electrochemical decomposition reactions on application of an electrical potential of, for example, 3 or 5 volts.

In addition of course, the sealing strand like the adhesive strip must be arranged along the entire pane edge between the glass panes in order to confine the electrochromic element jointly with the glass panes in a diffusion-impermeable manner. The sealing strand does not have to be flush with the pane edge. On the contrary, it can even overlap the pane edges, at least partially.

It can be advantageous if the electrochromic element is sealed towards the outside in known fashion with an additional sealant strand. Suitable for this purpose are in particular polysulfide-base materials used for double-glazing manufacture. This is especially appropriate if the electrochromic element is combined with at least one additional glass pane to form a double-glazing unit.

Preferably, the sealing strand adjoins directly the adhesive strip. It lies within the scope of the invention however to provide between the two components of the sealing element according to the invention at least one additional functional component, for example a separating layer or a primer layer, if the advantage achieved thereby is justified by the additional cost necessary. It is also possible for the glass panes in the region of the adhesive strip, the sealing strand or of an additional sealing strand to be provided with a primer in order to improve the adhesion of these materials to the glass.

The invention can be used to special advantage in the case of electrochromic elements with a polymer electrolyte layer which, in addition to the conduction salt necessary for provision of a sufficient quantity of cations (for example in accordance with WO 95-31 746 A1), incorporates at least one (meth)acrylic ester, at least one plasticizer and at least one polymerization initiator (EP 0 683 215 A1).

The invention is explained in further detail with the aid of the embodiments illustrated in the Figures. These show:

Figure 1 a first embodiment of the electrochromic element according to the invention in cross-section,

Figure 2 a second embodiment of the electrochromic element according to the invention in a similar representation.

The representation in the Figures is to be regarded as schematic. The dimensions are not to scale.

The electrochromic element according to Figure 1 consists of two 4 mm thick transparent float glass panes 1 and 2 which are each provided on the surfaces facing one another with transparent electrode layers 3, 4 of indium-tin oxide (ITO), of fluorine-doped tin oxide or another electrically conductive metal oxide. On electrode layer 3 is arranged an electrochromic layer 5 of tungsten oxide, whilst on electrode layer 4 is located an ion storage layer 6 of one or more metal oxides, such as for example cerium, vanadium, titanium, zirconium or nickel oxide. The edge area of the electrode layers 3, 4 is in each case uncoated over an area of a few millimeters, as can be seen in the Figure. Not illustrated are the busbars by means of which an electrical voltage is applied to the electrode layers 3, 4.

Between the coated glass panes is located a 0.9 mm thick, in situ polymerized polymer electrolyte layer 7 in accordance with EP 0 683 215 A1 with a conduction salt content in accordance with WO 95-31 746 A1. It adjoins directly a 0.9 mm thick by 9 mm wide transparent adhesive strip 8 of a low-crosslinked polyacrylate, which has been formed from an adhesive tape of Messrs 3M with the corresponding dimensions marketed under the commercial name Scotch Isotac VHB 4910, which had been subjected to a drying treatment before application. The polyacrylate of this adhesive strip possesses a glass transition temperature of between 5 and 10°C. The glass transition temperature was determined by means of Dynamic Thermomechanometry (see for example "Examination Methods in Chemistry", Georg Thieme Verlag, 2nd impression 1990, Chapter 1, page 13).

The surrounding groove remaining between the outer surface of the adhesive strip 8 facing towards the edge of the electrochromic element and the outer edges of the glass panes 1, 2 is filled with a sealing strand 9 of an epoxy sealant Araldit 2012. The sealing strand 9 is essentially flush with the outer edges of the glass panes 1, 2. It could however at least partially overlap these edges.

Figure 2 shows in cross-section an edge view of a double-glazing unit formed with an electrochromic unit according to the invention. The embodiment of the electrochromic element according to Figure 2 differs from that of Figure 1 in that the glass pane 1 possesses smaller dimensions than glass pane 2. As in the previous example, an adhesive strip 8 formed of a Scotch Isotac VHB 4910 adhesive tape of Messrs 3M is provided, this adjoining directly the polymer electrolyte layer 7. The width of the adhesive strip 8 at 6 mm is in this case chosen to be slightly less than in the foregoing example, in order to compensate at least partly for the loss of available vision area which is caused by the stepped design of the electrochromic element. The adhesive strip 8 is adjoined in this case by a sealing strand 9 formed of a butyl sealant, Bostik 5125. The step formed by the two glass panes 1 and 2 and by the outer face of the sealing strand 9 is filled with a further sealant strand 10 of a polysulfide double-glazing adhesive, for example of a polysulfide Naftotherm M 82 of Messrs Chemetall. The sealant strand 10 is only illustrated in section, whilst all other components of the double-glazing unit, such as for example the spacer and the at least one other glass pane, have been omitted entirely. The construction of double-glazing units is generally known, so that it is unnecessary to illustrate further details in this context.

The two electrochromic elements illustrated in the Figures have withstood various ageing tests without discernible damage.

Claims

1. Electrochromic element with an electrochromic arrangement enclosed between two plane substrates, which comprises at least two electrode layers, one electrochromic layer, one ion storage layer, and one polymer electrolyte layer formed in situ, where the polymer electrolyte layer adjoins a sealing element at the edge of the electrochromic element.

   characterized in that

   the sealing element consists of a plastically deformable liquid impermeable adhesive strip (8) of a polyacrylate, arranged between the two plane substrates (1, 2) and adjoining directly the polymer electrolyte layer (7), as well as of a sealing strand (9) adjacent thereto on the outside, consisting of a gas impermeable sealant chemically compatible with the adhesive strip (8).

2. Electrochromic element in accordance with Claim 1, **characterized in that** the adhesive strip (8) is formed of a polyacrylate tape.

3. Electrochromic element in accordance with Claim 1 or 2, **characterized in that** the adhesive strip (8) possesses a width of at least 5 mm.

4. Electrochromic element in accordance with Claim 3, **characterized in that** the adhesive strip(8) possesses a maximum width of 20 mm.

5. Electrochromic element in accordance with one of the foregoing claims, **characterized in that** the adhesive strip(8) consists of a polyacrylate with a maximum water content of 0.3 weight percent, preferably less than 0.05 weight percent.

6. Electrochromic element in accordance with one of the foregoing claims, **characterized in that** the adhesive strip (8) consists of a polyacrylate with a glass transition temperature below 10°C.

7. Electrochromic element in accordance with one of the foregoing claims, **characterized in that** the sealing strand (9) consists of a polyisobutylene or butyl rubber based butyl sealant.

8. Electrochromic element in accordance with Claim 7, **characterized by the fact that** the sealing strand (9) possesses a specific conductivity of less than 10^{-9} $\Omega^{-1}cm^{-1}$, preferably less than 10^{-11} $\Omega^{-1}cm^{-1}$ and a water vapour permeability according to DIN 53122-1.2 of less than 0.5 $g \cdot m^{-2} \cdot d^{-1}$.

9. Electrochromic element in accordance with one of Claims 1 to 6, **characterized in that** the sealing strand (9) consists of an epoxy sealant.

10. Electrochromic element in accordance with Claim 9, **characterized in that** the sealing strand (9) possesses a specific conductivity of less than 10^{-12} $\Omega^{-1}cm^{-1}$, preferably less than 10^{-13} Ω^{-1} cm^{-1} and a water vapour permeability according to DIN 53122-1.2 of less than 4.0 $g \cdot m^{-2} \cdot d^{-1}$.

11. Electrochromic element in accordance with one of the foregoing claims, **characterized in that** sealing strand (9) is adjoined by at least one further sealant strand (10), in particular one with polysulfide base.

12. Electrochromic element in accordance with one of the foregoing claims, **characterized in that** the polymer electrolyte layer (7) comprises at least one (meth)acrylic ester, at least one plasticizer and at least one polymerization initiator.

Abstract

The invention relates to an electrochromic element with an electrochromic arrangement that is enclosed between two planar substrates. Said arrangement comprises at least two electrode layers, an electrochromic layer, an ion storage layer and a polymer electrolyte layer which is formed in situ and which is adjacent to a sealing element towards the edge of the electrochromic element. According to the invention, the sealing element consists of a liquid proof, plastically deformable adhesive stripe (8) that is directly adjacent to the polymer electrolyte layer (7) and is arranged between the two substrates (1, 2). The adhesive stripe consists of a polyacrylate. A sealing line (9) made of a gastight sealant that is chemically compatible with the adhesive stripe is joined to the exterior of said adhesive stripe.

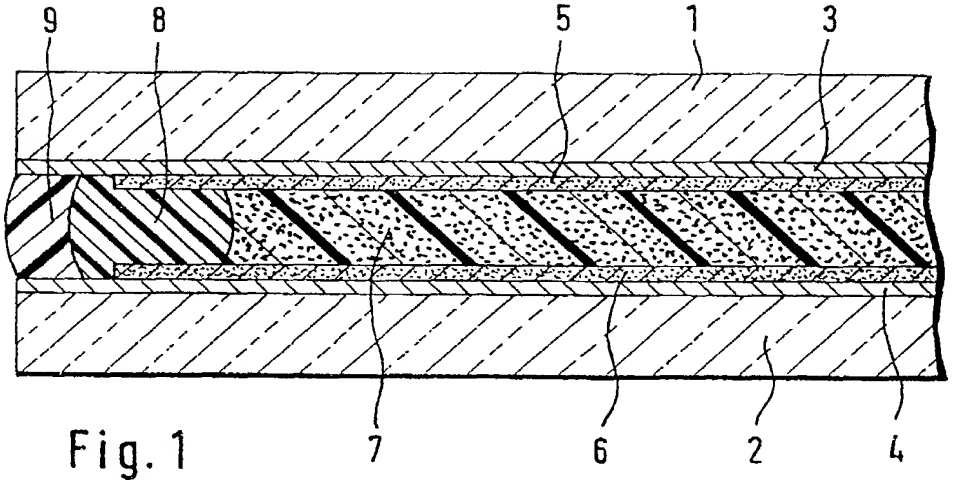

Fig. 1

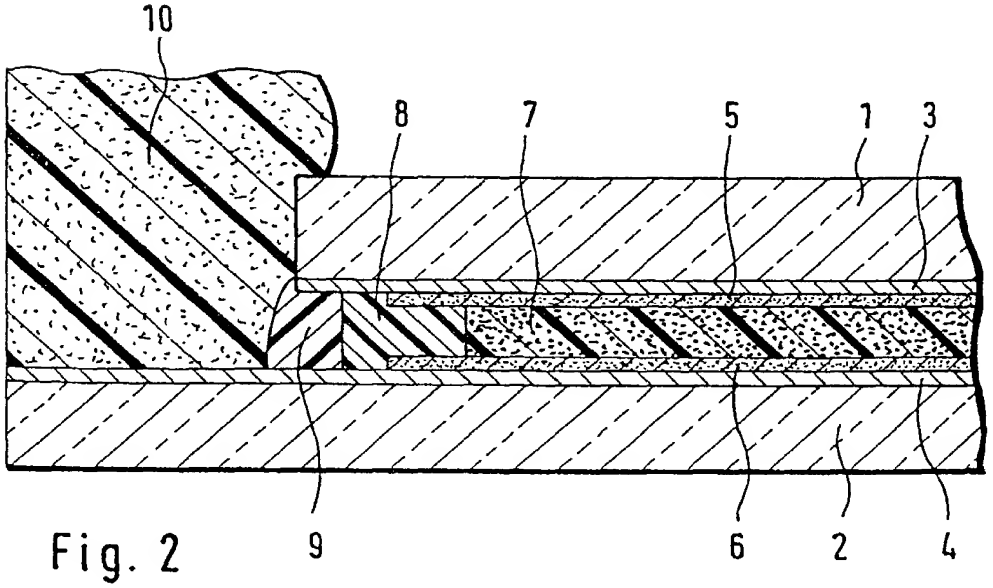

Fig. 2

09914082 .031102

Please type a plus sign (+) inside this box → [+]

Express Mail No. EL 469907515US

PTO/SB/81 (02-01)
Approved for use through 10/31/2002 OMB 0651-0035
U S Patent and Trademark Office, U S. DEPARTMENT OF COMMERCE
Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it display a valid OMB control number

POWER OF ATTORNEY OR AUTHORIZATION OF AGENT

Application Number	09/914,082
Filing Date	
First Named Inventor	Dirk Joedicke
Title	Electrochromic Element
Group Art Unit	
Examiner Name	
Attorney Docket Number	1-15478

I hereby appoint:

☐ Practitioners at Customer Number [ ] → *Place Customer Number Bar Code Label here*

OR

☒ Practitioner(s) named below:

Phillip S. Oberlin, Reg.No. 19,066; Donald A. Schurr, Reg.No. 34,247; D. Edward Dolgorukov, Reg.No. 26,266; Mark A. Hixon, Reg. No. 44,766; Stephen P. Evans, Reg.No. 47,281; and Angelica M. Colwell, Reg.No. 46,637

all of the firm of Marshall & Melhorn, LLC
Four SeaGate - 8th Floor
Toledo, Ohio 43604 USA

as my/our attorney(s) or agent(s) to prosecute the application identified above, and to transact all business in the United States Patent and Trademark Office connected therewith.

Please change the correspondence address for the above-identified application to:

☐ The above-mentioned Customer Number.

OR

☐ Practitioners at Customer Number [ ] → *Place Customer Number Bar Code Label here*

OR

☐ Firm *or* Individual Name					
Address					
Address					
City		State		Zip	
Country					
Telephone		Fax			

I am the:

☐ Applicant/Inventor.

☒ Assignee of record of the entire interest. See 37 CFR 3.71.
Statement under 37 CFR 3.73(b) is enclosed. (Form PTO/SB/96).

SIGNATURE of Applicant or Assignee of Record

Name	FLABEG GMBH & CO. KG	Title: authorized represent.
Signature	X [signature]	Typed Name: Dr. Hartmut Wittkopf
Date	X Furth im Wald, 02.07.2002 July 2, 2002	Head of Research and Development

NOTE: Signatures of all the inventors or assignees of record of the entire interest or their representative(s) are required. Submit multiple forms if more than one signature is required, see below*.

☐ *Total of _____ forms are submitted.

COMBINED DECLARATION AND POWER OF ATTORNEY
IN ORIGINAL APPLICATION

ATTORNEY DOCKET NO. 1-15478

As a below named inventor, I hereby declare that:

My residence, post office address, and citizenship are as stated below next to my name,

I believe that I am the original, first, and sole inventor (if only one name is listed below) or an original, first, and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

ELECTROCHROMIC ELEMENT

the specification of which

(check one)

___ is attached hereto.

XX is a filing under 35 USC 371 of PCT International Application No. PCT/DE00/00553 filed 24 February 2000.

XX was filed on _____ as U.S. Serial No. 09/914,082 and was amended on 22 August 2001.

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information, which is material to patentability as defined in 37, Code of Federal Regulations, § 1.56,

X and which is material to the examination of this application, namely, information where there is a substantial likelihood that a reasonable Examiner would consider it important in deciding whether to allow the application to issue as a patent; and

___ if this is a continuation-in-part application, information that occurred between the filing date of the prior application(s) and the national or PCT international filing date of this application, in accordance with 37 CFR 1.63(e); and

___ in compliance with this duty, there is attached an information disclosure statement, in accordance with 37 CFR 1.98.

I hereby claim foreign priority benefits under Title 35, United States Code, §119 of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed:

Prior Foreign Application(s)			Priority Claimed	
199 08 737.7	Germany	01 March 1999 ✓	XX	
(Number)	(Country)	(Day/Month/Year Filed)	Yes	No
(Number)	(Country)	(Day/Month/Year Filed)	Yes	No

I hereby claim the benefit under Title 35, United States Code, §120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code, §112, I acknowledge the duty to disclose material information as defined in Title 37, Code of Federal Regulations, §1.56 which became available between the filing date of the prior application and the national or PCT international filing date of this application:

(Application No.)	(Filing Date)	(Status) (patented, pending, abandoned)
(Application No.)	(Filing Date)	(Status) (patented, pending, abandoned)

I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith with full power of substitution and revocation: Phillip S. Oberlin, Reg. No. 19,066; D. Edward Dolgorukov, Reg. No. 26,266; Donald A. Schurr, Reg. No. 34,247; Mark A. Hixon, Reg. No. 44,766; Stephen P. Evans, Reg. No. 47,281; and Angelica M. Colwell, Reg. No. 46,637. Address all telephone calls to Phillip S. Oberlin at telephone number (419) 249-7149. Address all correspondence to MARSHALL & MELHORN, LLC, Four Seagate - 8th Floor, Toledo, Ohio 43604, Attention: Phillip S. Oberlin.

5-

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

1-00 Full name of sole or first inventor DIRK JOEDICKE

Inventor's signature [signature] Date 07.11.2001
November 7, 2001

Residence Koehlerweg 4B, D-93437 Furth im Wald, Germany DEX

Citizenship Germany Post Office Address same as above

2-00 Full name of second joint inventor, if any HANS-JOACHIM BECKER

Second inventor's signature [signature] Date 9.11.2001
November 9, 2001

Residence Himmelackerring 10, D-91459 Markt Erlbach, Germany DEX

Citizenship Germany Post Office Address same as above

3-00 Full name of third joint inventor, if any VOLKER GUMPRICH

Third inventor's signature [signature] Date 30.10.2001
October 30, 2001

Residence Berghäuschensweg 242, D-41468 Neuss, Germany DEX

Citizenship Germany Post Office Address same as above

4-00 Full name of fourth joint inventor, if any RICHARD BATCHELOR

Fourth inventor's signature X R.A. Batchelor Date X 6th December 2001
December 6, 2001

Residence 39 Blundell Drive Southport, Merseyside PR8 4RE Great Britain GBX

Citizenship Great Britain Post Office Address same as above

Full name of fifth joint inventor, if any _____

Fifth inventor's signature _____ Date _____

Residence _____

Citizenship _____ Post Office Address _____

Full name of sixth joint inventor, if any _____

Sixth inventor's signature _____ Date _____

Residence _____

Citizenship _____ Post Office Address _____